

Engineered spin phase diagram of two interacting electrons in semiconductor nanowire quantum dots

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Spin properties of two interacting electrons in a quantum dot (QD) embedded in a nanowire with controlled aspect ratio and longitudinal magnetic fields are investigated by using a configuration interaction (CI) method and exact diagonalization (ED) techniques. The developed CI theory based on a three-dimensional (3D) parabolic model provides explicit formulations of the Coulomb matrix elements and allows for straightforward and efficient numerical implementation. Our studies reveal fruitful features of spin singlet-triplet transitions of two electrons confined in a nanowire quantum dot (NWQD), as a consequence of the competing effects of geometry-controlled kinetic energy quantization, the various Coulomb interactions, and spin Zeeman energies. The developed theory is further employed to study the spin phase diagram of two quantum-confined electrons in the regime of “cross over” dimensionality, from quasi-two-dimensional (disk-like) QDs to finite one-dimensional (rod-like) QDs.

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I. INTRODUCTION

Stimulated by recent success in coherent control of two-electron spin in laterally coupled quantum dots (QDs),¹ the spin states of two interacting electrons in semiconductor QDs have received increasingly considerable attention. Accessible and engineerable spin states of few electrons in QDs thus have become one of the basic features required by the quantum information applications in which electron spins are utilized as quantum bit.^{2,3} For two-dimensional (2D) epitaxial QDs, magnetic field induced spin singlet-triplet (ST) transitions of two-electron ground states have been studied extensively for years.^{4,5,6,7} The underlying physics of the ST transitions is usually associated with the energetic competition between quantized kinetic energies, the coulomb interactions, and spin Zeeman energies. Reversely switching the singlet and triplet spin states of a lateral two-electron QD is feasible by utilizing electrical control.⁸ Moreover, it has been both theoretically and experimentally shown that more complex oscillating spin phases can be generated either by reducing the lateral confinement or by increasing an applied magnetic field.^{9,10,11,12}

Recently, the local-gate electrical depletion^{13,14,15} and the bottom-up grown techniques^{16,17} have been developed for the fabrication of few-electron QDs embedded in a nanowire. These experimental developments open up an opportunity of exploring the *cross over* mechanisms from the 2D (disk-like) to the finite 1D (rod-like) QD regimes. Such nanowire quantum dots (NWQDs) are advantageous for geometrical control over a wide range of aspect ratio a (typically from $a \sim 10^{-1}$ to $a \gg 1$).^{16,17} The excellent versatility of shape and dimensionality makes NWQDs a suitable nanomaterial for scalable quantum electronics. Very recently, successful fabrication of single electron transistors made of InAs based gate-defined NWQDs and observations of the singlet-triplet transi-

tions of two electrons in the QDs have been demonstrated.¹⁸ How the highly tunable longitudinal confinement of NWQD affects and can be utilized to tailor the spin properties of few electrons in NWQDs are interesting subjects worth studying.

The above experimental efforts motivate us to perform a theoretical investigation of the spin states of two electrons in InAs-based NWQDs¹⁸ by using a developed configuration interaction (CI) theory and exact diagonalization techniques.¹⁹ The developed CI theory is based on the 3D parabolic model with arbitrary transverse and longitudinal confinement strengths^{20,21} and provides explicit generalized formulations of the Coulomb matrix, and thus allows for straightforward and efficient numerical or even semi-analytical implementation widely applicable for various cylindrically symmetric QDs. Our exact diagonalization studies of two-electron charged NWQDs with controlled geometric aspect ratios and longitudinal magnetic fields reveal fruitful features of spin singlet-triplet transitions, as a consequence of the competing effects of geometry-engineered kinetic energy quantization, the various Coulomb interactions, and spin Zeeman energies. The developed theory is further employed to study the spin phase diagram of two quantum-confined electrons in the regime of “cross over” dimensionality from quasi-2D (disk-like) QDs to finite 1D (rod-like) QDs.

This article is organized as follows: Section II describes the theoretical model and the developed configuration interaction theory for few-electron problems of three-dimensionally confining quantum dots. In Sec. III, we present and discuss the calculated results of magneto-energy spectrum, the ST transitions and geometry-engineered spin phase diagrams of two-electron charged quantum dots embedded in nanowires. Concluding remarks are presented in Sec. IV.

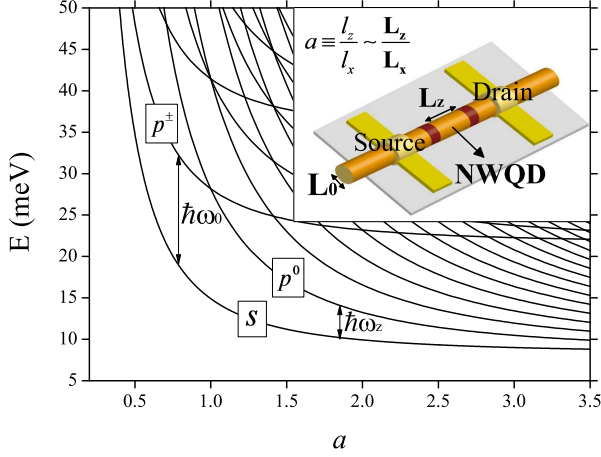


FIG. 1: (Color online) Single-electron energy spectrum as a function of aspect ratio a of a NWQD with fixed lateral confinement $\hbar\omega_0 = 13.3$ meV at zero magnetic field obtained from the 3D parabolic model. The considered lateral confinement strength $\hbar\omega_0 = 13.3$ meV corresponds to the cross section diameter $\mathbf{L}_0 \sim 65$ nm for a cylindrical InAs nanowire. The low-lying s -, p^\pm -, and p^0 -orbitals are relevant to a two-electron problem. The energy quantization for a short (long) NWQD with $a < 1$ ($a > 1$) is characterized by the energy difference between the lowest and first excited orbitals $\hbar\omega_0$ ($\hbar\omega_z$).

II. MODEL

A. Single-particle model

We begin with the problem of a single electron in a NWQD with a uniform longitudinal magnetic field $\mathbf{B} = (0, 0, B)$, which is described by the single-electron Hamiltonian,

$$H_0 = \frac{1}{2m^*}(\mathbf{p} + e\mathbf{A})^2 + V_c(x, y, z) + H_Z. \quad (1)$$

Here the first term indicates the term of kinetic energy with $\mathbf{A} = (B/2)(-y, x, 0)$ being the vector potential in symmetric gauge, m^* the effective mass of electron and e the charge of an electron. The second term is the confining potential of NWQD modeled by

$$V_c(x, y, z) = \frac{1}{2}m^*[\omega_0^2(x^2 + y^2) + \omega_z^2z^2] \quad (2)$$

with ω_0 and ω_z parametrizing, respectively, the transverse and the longitudinal confining strength. The last term is the spin Zeeman energy $H_Z = g^*\mu_B Bs_z$, in terms of the z -component of electron spin $s_z = \pm 1/2$, the effective Lande g -factor of electron g^* and the Bohr magneton μ_B . The single-particle Hamiltonian (1) leads to the extended Fock-Darwin single-particle spectrum

$$\begin{aligned} \epsilon_{n,m,q,s_z} = & \hbar\omega_+ \left(n + \frac{1}{2}\right) + \hbar\omega_- \left(m + \frac{1}{2}\right) \\ & + \hbar\omega_z \left(q + \frac{1}{2}\right) + E_Z \end{aligned} \quad (3)$$

where $n, m, q = 0, 1, 2, \dots$ denote oscillator quantum numbers, $E_Z = g^*\mu_B Bs_z$ is the spin Zeeman energy, $\omega_\pm = \omega_h \pm \omega_c/2$ are in terms of the hybridized frequency $\omega_h \equiv (\omega_0^2 + \omega_c^2/4)^{1/2}$ and the cyclotron frequency $\omega_c = eB/m^*$. The corresponding eigenstate $|n, m, q\rangle$ possesses the orbital angular momentum projection $\ell_z = \hbar(n - m)$ and the parity $P = 1$ ($P = -1$) with respect to z -axis for an even (odd) q number. The wave function of the lowest orbital is given by

$$\begin{aligned} \psi_{000}(\mathbf{r}) = & \left[(2\pi)^{3/4}l_h\sqrt{l_z}\right]^{-1} \\ & \times \exp\left[-\frac{1}{4}\left(\frac{x^2 + y^2}{l_h^2} + \frac{z^2}{l_z^2}\right)\right], \end{aligned} \quad (4)$$

with the characteristic lengths of the wave function extents $l_h = \sqrt{\hbar/2m^*\omega_h}$ and $l_z = \sqrt{\hbar/2m^*\omega_z}$. The wave functions of other excited states can be generated by successively applying the following defined raising operators¹⁹

$$\begin{aligned} a^\dagger &= \frac{1}{\sqrt{2}}\left[\frac{x + iy}{2l_h} - l_h(\partial_x + i\partial_y)\right], \\ b^\dagger &= \frac{1}{\sqrt{2}}\left[\frac{x - iy}{2l_h} - l_h(\partial_x + i\partial_y)\right], \\ a_z^\dagger &= \frac{z}{2l_z} - l_z\partial_z \end{aligned} \quad (5)$$

onto the ground state $|0, 0, 0\rangle$, i.e.

$$|n, m, q\rangle = \frac{(\hat{a}^\dagger)^n(\hat{b}^\dagger)^m(\hat{a}_z^\dagger)^q}{\sqrt{n!m!q!}}|0, 0, 0\rangle. \quad (6)$$

The diameter of cross section of bottom-up synthesized nanowire is typically $\sim 50 - 70$ nm. By contrast, the length of a QD in a nanowire, defined by imposed electrodes or heterostructure potential barriers, is highly tunable over a wide range from 10 to 300 nm.¹⁷ For characterizing the geometry of a NWQD, it is convenient to define the parameter of aspect ratio,

$$a \equiv \frac{l_z}{l_0} = \sqrt{\frac{\omega_0}{\omega_z}} \quad (7)$$

according to the characteristic length of the lowest orbital wave function based on the 3D parabolic model. A rod-like (disk-like) NWQD is characterized by the value of aspect ratio $a > 1$ ($a < 1$), where the longitudinal extent of the electron wave function is longer (shorter) than the transverse one on the cross section of the nanowire. Notably, the effective aspect ratio $a = l_z/l_0$ defined here is not but very close to the geometric aspect ratio a_{geom} , namely $a \simeq a_{\text{geom}} = \mathbf{L}_z/\mathbf{L}_0$ (\mathbf{L}_z) being the cross section diameter (length) of NWQD.

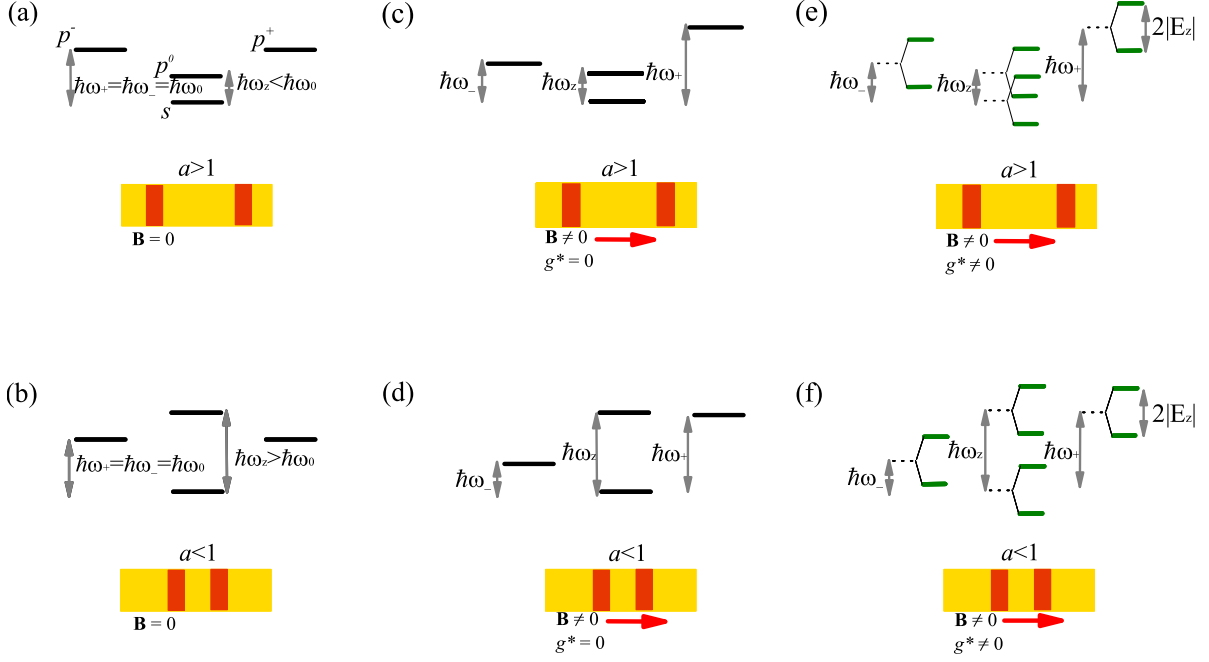


FIG. 2: (Color online) Schematic illustration of the electronic structures, consisting of few relevant low lying orbitals (one s - and three p -orbitals), of long rod-like NWQDs [(a)(c)(e)] and short disk-like NWQDs [(b)(d)(f)] with or without longitudinal magnetic field B and including or excluding the spin Zeeman splitting E_z ($g^* = 0$ or $g^* \neq 0$). (a) $a > 1$ and $B = 0$; (b) $a < 1$ and $B = 0$; (c) $a > 1$, $B \neq 0$ and $g^* = 0$; (d) $a < 1$, $B \neq 0$ and $g^* = 0$; (e) $a > 1$, $B \neq 0$ and $g^* \neq 0$; (f) $a < 1$, $B \neq 0$ and $g^* \neq 0$.

Figure 1 presents the calculated single-electron energy spectrum as a function of aspect ratio a for a NWQD with fixed lateral confinement $\hbar\omega_0 = 13.3$ meV at zero magnetic field according to Eq.(3). The chosen parameter of lateral confinement $\hbar\omega_0 = 13.3$ meV is determined by fitting the numerically calculated energy separation between the two lowest single-electron orbitals of a cylindrical InAs/InP NWQD of cross section diameter $L_0 = 65$ nm by 3D finite difference simulation. In the simulation, the Schrödinger equation for a single electron confined in a 3D cylindrical potential well is solved by using finite difference method, with the used parameters: the effective mass $m^* = 0.023m_0$ of electron for InAs and the InAs/InP band edge offset $V_b = 0.6$ eV as the barrier height of the confining potential.^{17,22}

In a two-electron (2e) problem, the most relevant orbitals are the two lowest ones because the kinetic energy difference between the two orbitals is the main energy cost, in competition with the coulomb or spin Zeeman energies, for a spin triplet state to be the ground state of two-electron. By convention, we from now on name the lowest single electron state $|n, m, q\rangle = |0, 0, 0\rangle$ as s -orbital, and the next three p -shell states $|0, 0, 1\rangle$, $|1, 0, 0\rangle$, and $|0, 1, 0\rangle$ as p^0 -, p^+ -, and p^- -orbitals, respectively. According to Eq. (3), the energy of the lowest s -orbital is explicitly given by

$$\epsilon_{s,s_z} = \frac{1}{2} (\hbar\omega_+ + \hbar\omega_- + \hbar\omega_z) + g^* \mu_B B s_z, \quad (8)$$

and those of the three p -shell orbitals are respectively given by

$$\begin{aligned} \epsilon_{p^0,s_z} &= \epsilon_{s,s_z} + \hbar\omega_z, \\ \epsilon_{p^+,s_z} &= \epsilon_{s,s_z} + \hbar\omega_+, \\ \epsilon_{p^-,s_z} &= \epsilon_{s,s_z} + \hbar\omega_-. \end{aligned} \quad (9)$$

For $B = 0$, we have $\epsilon_{s,s_z} = \hbar\omega_0 (1 + 1/2a^2)$, $\epsilon_{p^0,s_z} = \epsilon_{s,s_z} + \hbar\omega_0/a^2$, and $\epsilon_{p^+,s_z} = \epsilon_{p^-,s_z} = \epsilon_{s,s_z} + \hbar\omega_0$ according to Eqs.(7) and (9). Here, the p^+ - and p^- -orbitals are degenerate with the same energy separation from the s -orbital, $\hbar\omega_{\pm} = \hbar\omega_0$, while the p^0 -orbital is energetically higher than s -orbital by $\hbar\omega_z = \hbar\omega_0/a^2$. Obviously, p^0 (p^{\pm}) is the second lowest orbital for a long (short) NWQD with $a > 1$ ($a < 1$) at zero magnetic field, as shown in Fig. 1. For a symmetric NWQD with $a = 1$, the p^0 - and p^{\pm} -orbitals form a 3-fold orbital-degenerate shell. Figure 2 (a) [(b)] schematically depicts the low-lying orbitals of a long [short] NWQD with $a > 1$ [$a < 1$] at zero magnetic field.

Applying a longitudinal magnetic field onto a cylindrical NWQD breaks the degeneracy of p^+ - and p^- -orbitals. The orbital Zeeman effect lowers (raises) the energy level of the p^- (p^+)-orbital from $\hbar\omega_0$ to $\hbar\omega_-$ ($\hbar\omega_+$). Thus, if a long NWQD is subjected to a sufficiently strong magnetic field, the second lowest orbital of the dot could be changed from the p^0 to p^- . By contrast, the second lowest orbital of a short NWQD is always the p^- -orbital.

Therefore, the characteristic energy quantization of the p^- -orbitals, $\hbar\omega_- = \hbar(\omega_0^2 + \omega_c^2/4)^{1/2} - \hbar\omega_c/2$, is often a key parameter for a short NWQD or a moderately long NWQD with strong magnetic field. Considering wide-band gap materials such as GaAs, the g -factors are usually small and the spin Zeeman effect on the energy shift of orbital is negligible. Figure 2(c) [(d)] depicts the B -dependent electronic orbitals of a long [short] NWQDs, where vanishing spin Zeeman splitting is assumed ($g^* = 0$ is set).

For a low energy gap material with larger g^* , like InAs, the spin Zeeman effect could be significant in the spin ST transition of two-electron QD. Figure 2(e) [(f)] schematically shows the spin-resolved electronic orbitals of a long [short] NWQDs with $B \neq 0$ and $g^* \neq 0$ by the spin Zeeman splitting $2E_Z$. With the spin Zeeman effect, all the spin-up (spin-down) orbitals are energetically lowered (raised) by $E_Z = g^*\mu_B B/2$ according to Eq. (3). If the applied magnetic field or the g -factor of material is so large that the spin Zeeman splittings exceed the kinetic energy quantization of QD, both of the two lowest single-electron states are the spin-up ones and the ground state of the 2e dot is ensured to be a spin triplet state simply according to spin Pauli exclusion principle.

In this work, the following formulation for the g -factor of an InAs-based QD is adopted^{17,23}

$$g^* = g \left[1 - \frac{P^2}{3} \frac{\Delta_{\text{SO}}}{E_g^{\text{eff}} (E_g^{\text{eff}} + \Delta_{\text{SO}})} \right], \quad (10)$$

where E_g^{eff} is the effective energy gap of semiconductor QD, $g = 2.0$ is the Lande g -factor for free electron, Δ_{SO} is the spin-orbit splitting in the valence band, and P is the parameter of interband transition matrix element.²³

Here, the effective energy gap of a QD can be estimated as $E_g^{\text{eff}} = E_g^{\text{bulk}} + \epsilon_{s,s_z}$, where E_g^{bulk} is the bulk energy gap and ϵ_{s,s_z} is the quantization energy of the

lowest electronic orbital of the QD with $B = 0$ measured from the conduction band edge. For InAs-based QDs, we take the following parameter values: $E_g^{\text{bulk}} = 460$ meV, $\Delta_{\text{SO}} = 390$ meV, $P^2 = 21.5$ eV.¹⁷ Accordingly, the value of g^* for a symmetric NWQD with $\mathbf{L}_0 = \mathbf{L}_z = 65$ nm is estimated as large as $g^* \approx -11$.¹⁷

B. Interacting few-electron model

The interacting Hamiltonian of few electrons in a NWQD can be expressed in the form of second quantization as

$$H = \sum_{i,\sigma} \epsilon_{i\sigma} c_{i\sigma}^\dagger c_{i\sigma} + \frac{1}{2} \sum_{ijkl,\sigma\sigma'} \langle ij|V|kl \rangle c_{i\sigma}^\dagger c_{j\sigma'}^\dagger c_{k\sigma'} c_{l\sigma}, \quad (11)$$

where i, j, k, l denote the composite indices of single electron orbitals such as $|i\rangle = |n_i, m_i, q_i\rangle$, $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) the electron creation (annihilation) operators, and $\sigma = \pm$ the electron spins $s_z = \pm \frac{1}{2}$. The first (second) term on the right hand side of Eq.(11) represents the kinetic energy of electrons (the Coulomb interactions between electrons) and the Coulomb matrix elements are defined as

$$\langle ij|V|kl \rangle \equiv \frac{e^2}{4\pi\kappa} \int \int d\mathbf{r}_1 d\mathbf{r}_2 \psi_i^*(\mathbf{r}_1) \psi_j^*(\mathbf{r}_2) \times \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} \psi_k(\mathbf{r}_2) \psi_l(\mathbf{r}_1), \quad (12)$$

where κ is the dielectric constant of dot material. For InAs material, we take $\kappa = 15.15$. After lengthy derivation, one can obtain the generalized Coulomb matrix elements for the case of $a \geq 1$:

$$\begin{aligned} \langle n_i m_i q_i; n_j m_j q_j | V | n_k m_k q_k; n_l m_l q_l \rangle = & \left(\frac{1}{\pi l_h} \right) \frac{\delta_{R_L, R_R} \cdot \delta_{q_i+q_j+q_l+q_k, \text{even}}}{\sqrt{n_i! m_i! q_i! n_j! m_j! q_j! n_k! m_k! q_k! n_l! m_l! q_l!}} \\ & \times \sum_{p_1=0}^{\min(n_i, n_l)} \sum_{p_2=0}^{\min(m_i, m_l)} \sum_{p_3=0}^{\min(q_i, q_l)} \sum_{p_4=0}^{\min(n_j, n_k)} \sum_{p_5=0}^{\min(m_j, m_k)} \sum_{p_6=0}^{\min(q_j, q_k)} p_1! p_2! p_3! p_4! p_5! p_6! \\ & \times \binom{n_i}{p_1} \binom{n_l}{p_1} \binom{m_i}{p_2} \binom{m_l}{p_2} \binom{q_i}{p_3} \binom{q_l}{p_3} \binom{n_j}{p_4} \binom{n_k}{p_4} \binom{m_j}{p_5} \binom{m_k}{p_5} \binom{q_j}{p_6} \binom{q_k}{p_6} \\ & \times (-1)^{u+v/2+n_j+m_j+q_j+n_k+m_k+q_k} \left(\frac{1}{2} \right)^u x^{u+1/2} \\ & \times \frac{\Gamma(\frac{1+2u+v}{2}) \Gamma(1+u) \Gamma(\frac{1+v}{2})}{\Gamma(\frac{3+2u+v}{2})} {}_2F_1 \left(1+u, \frac{1+2u+v}{2}; \frac{3+2u+v}{2}; 1-x \right), \end{aligned} \quad (13)$$

where we define $u = m_i + m_j + n_l + n_k - (p_1 + p_2 + p_4 + p_5)$, $v = (q_i + q_l + q_j + q_k) - 2(p_3 + p_6)$, $R_L = (m_i + m_j) -$

$(n_i + n_j) = -(\ell_{z,i} + \ell_{z,j})$, $R_R = (m_l + m_k) - (n_l + n_k) =$

$-(\ell_{z,l} + \ell_{z,k})$, $x \equiv \omega_z/\omega_h$, and ${}_2F_1$ is the hypergeometric function. The δ -functions $\delta_{q_i+q_j+q_l+q_k, \text{even}}$ and δ_{R_L, R_R} in the formulation ensure the conservation of the parity with respect to z -axis and the z -component of angular momentum of system L_z , respectively. The formulation of Eq. (13) is confirmed by computing the Coulomb integral numerically.

For short NWQDs with $a < 1$, the formulations of the Coulomb matrix elements are obtained by simply taking Euler's hypergeometric transformation for the hypergeometric function in Eq. (13), i.e., replacing

$${}_2F_1\left(1+u, \frac{1+2u+v}{2}; \frac{3+2u+v}{2}; 1-x\right)$$

by

$$x^{-\frac{1+2u+v}{2}} {}_2F_1\left(\frac{1+v}{2}, \frac{1+2u+v}{2}; \frac{3+2u+v}{2}; 1-\frac{1}{x}\right).$$

The generalized formulations for the Coulomb matrix elements based on the 3D asymmetric parabolic model are probably for the first time derived, which allows for straightforward implementation of the CI theory and is widely applicable to arbitrary 3D confining semiconductor nanostructures.

C. Exact diagonalization

Based on the CI theory presented above, we follow the standard numerical exact diagonalization procedure to calculate the energy spectrum of N_e interacting electrons in a NWQD.¹⁹ The numerically exact results are obtained by increasing the numbers of chosen single electron orbital basis and the corresponding N_e -electron configurations until a numerical convergence is achieved. In the full configuration interaction (FCI) calculation for a 2e problem, we usually take the number of single electron orbitals typically from 20 to 26 and that of the corresponding 2e configurations from 190 to 325 to have a satisfactory numerical convergence.

III. NUMERICAL RESULTS AND DISCUSSION

A. Magnetic-field induced ST transitions

Let us first consider two interacting electrons in a rod-like NWQD with the aspect ratio $a = 3$ and the transverse confining strength $\hbar\omega_0 = 13.3$ meV using FCI calculation. The low-lying magneto-energy spectrum of the two-electron NWQD is shown in Fig. 3, which consists of a spin singlet state branch, labeled by S, and three triplet state branches split by the spin Zeeman energy, labeled by $T_{L_0}^+$, $T_{L_0}^0$ and $T_{L_0}^-$ according to the z -component of total spin ($S_z = +1$, $S_z = 0$ and $S_z = -1$), respectively.^{18,24} Since usually only triplet states with $S_z = +1$ are involved in ST transitions, we shall use $T_{L|L_z|}$ to

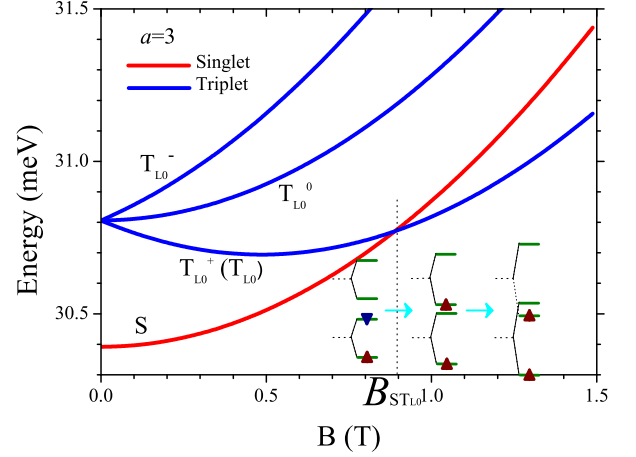


FIG. 3: (Color online) Magneto-energy spectrum of two interacting electrons in a NWQD with transverse confining strength $\hbar\omega_0 = 13.3$ meV and aspect ratio $a = 3$.

denote the triplet states with angular momentum L_z through out this article, skipping the superscript $+$ of $T_{L|L_z|}^+$ for brevity.

The main configurations of the two-electron ground states around the critical magnetic field are schematically shown in the lower right corner of Fig. 3. In the weak magnetic field regime ($B < B_{STL_0} \sim 0.9$ T), the two electrons in the NWQD mainly occupy the lowest s -orbital simply following the Aufbau principle, and form a spin singlet ground state. With increasing B , the triplet state T_{L_0} is more energetically favorable than the singlet state because of the increasing spin Zeeman energy, the reduced Coulomb repulsion, and exchange energy between the two spin polarized electrons. A crossing of the singlet branch and the triplet state branch T_{L_0} is observed at the critical magnetic field $B_{STL_0} = 0.9$ T. Such magnetic-field induced ST transitions are attributed to the energetic competition between single particle energy quantization, the spin Zeeman energy, and the various Coulomb interactions including the direct, exchange, and correlation interactions as well.¹⁰

Other weak spin-related terms, such as the spin-orbital coupling (SOC) with 1-2 order of magnitude smaller than the kinetic quantization of QD are neglected in the Hamiltonian of Eq.(11). The SOC mixes the spin of the S and T_{L_0} states and creates an anti-crossing of the S- and T_{L_0} -branches around the B_{ST} with a small energy gap, typically only $\sim 0.1 - 0.5$ meV as observed in previous experiments.¹⁸

B. Spin phase diagram

Figure 4 shows the calculated spin phase diagrams of the two-electron ground state of the NWQDs with a fixed cross section diameter (fixed $\hbar\omega_0 = 13.3$ meV) but various lengths (various $\hbar\omega_z$) with respect to the applied

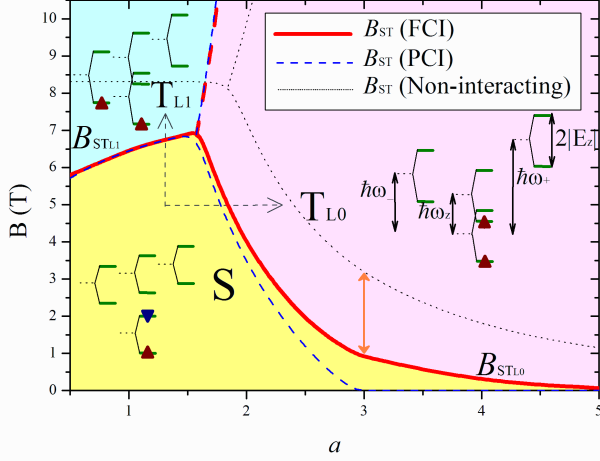


FIG. 4: (Color online) Spin phase diagrams of two-electron NWQDs of lateral confinement $\hbar\omega_0 = 13.3$ meV with respect to tunable magnetic field B and aspect ratio a . The phases are distinguished by the curves of critical magnetic field B_{ST} obtained from non-interacting (black dotted), PCI (blue dashed), and FCI (red solid) calculations.

magnetic field B and the aspect ratio a . Three phases (S, T_{L0} , and T_{L1}) are distinguished by the curves of critical magnetic field B_{ST} in Fig. 4. Correspondingly, the main configurations of the 2e ground states are depicted inside the colored regions of the phases. To identify the various underlying mechanisms in the phase diagrams, including the spin Zeeman effect and the inter-particle Coulomb interactions, the spin phase diagrams are calculated by using non-interacting, full CI, and partial CI calculations, respectively.

In the non-interacting calculation, the coulomb interactions are artificially disabled and the considered ST transitions are induced only by the spin Zeeman effect. The comparison between the results of non-interacting and FCI calculations allows us to distinguish effects of the Coulomb interaction and spin Zeeman coupling on the ST transitions. In particular, to highlight the Coulomb correlation effect, a partial configuration interaction (PCI) calculation is also performed for the spin phase diagrams, in which only the lowest energy configuration is taken as the sole basis and the couplings from higher energy configurations are excluded.

The essential features of the phase diagrams can be realized based on the non-interacting picture. For a not very long (small or moderate a) NWQD with weak B , the 2e ground state is likely to be the spin singlet state S, simply following Aufbau principle (the yellow region in Fig. 4). Starting from the singlet phase S, the two-electron ground state of a NWQD might be switched to the spin triplet phases (the pink region T_{L0} or the cyan region T_{L1}) by increasing either a or B (see the horizontal and vertical dashed lines with arrows in Fig. 4 for the guidance of eyes).

Following the horizontal dashed line, the longitudinal

energy quantization $\hbar\omega_z$ is decreased by the increase of a . With the addition of spin Zeeman term, the spin-up level of p^0 -orbital could become even lower than the spin-down level of s -orbital if the decreasing $\hbar\omega_z$ is so small as that $\hbar\omega_z < 2|E_z|$ (see the difference between the schematic configurations for the S and T_{L0} states). In this situation, the 2e ground state can transit to the spin triplet states T_{L0} , simply following spin Pauli exclusion principle. On the other hand, the transition of a 2e ground state of NWQD from the singlet state S to the triplet one T_{L1} is shown also possible by increasing the strength of applied magnetic field. Following the vertical dashed line, increasing B reduce the energy separation between the s - and p^- -orbital levels, i.e. $\hbar\omega_-$. Similar to the case of S- T_{L0} transition, a S- T_{L1} transition can happen as the decreased $\hbar\omega_-$ is so small as that $\hbar\omega_- < 2|E_z|$. In the non-interacting picture, $B_{ST_{L0}}$ is explicitly given by $B_{ST_{L0}} = \hbar\omega_0/g^*\mu_B a^2$, showing a quadratic decay with a , while the critical magnetic field $B_{ST_{L1}}$ for S- T_{L1} transitions is dependent only on $\hbar\omega_0$ and remains nearly constant in the $B - a$ plot.

The Coulomb interactions are shown to reduce the singlet phase area in the diagrams from the comparison between the non-interacting and CI results. For example, the segment of vertical solid line at $a = 3$ in Fig. 4 indicates that the critical magnetic field is significantly reduced from $B_{ST_{L0}}(\text{Non-interacting}) = 3.2$ T to $B_{ST_{L0}}(\text{FCI}) = 0.9$ T as the Coulomb interactions are taken into account. This is because the spin triplet states gain additional negative exchange energies while the singlet state does not. We also notice that the $B_{ST_{L1}}$ for the S- T_{L1} transition no longer remains constant but slightly increases with increasing a because the strength of the coulomb interactions is reduced by the increase of dot volume.

Basically, the results obtained from the FCI and PCI calculations have similar features except for those in the regime of high a ($a > 3$). While the PCI calculation shows the vanishing $B_{ST_{L0}}$ for $a \sim 3$, the FCI calculation yields the always non-zero $B_{ST_{L0}}$. This means that the Coulomb correlations energetically favor the spin singlet state as ground state and become more pronounced in long NWQDs.

C. Crossover from disk-like to rod-like QDs

The spin phase diagrams of Fig. 4 suggest that purposely accessing a specific spin phase of two-electron is feasible through the geometrical control of NWQDs. For instance, the ground state of a two-electron NWQD can be switched from the singlet S to the triplet state T_{L0} by increasing the aspect ratio a at the fixed $B = 5$ T (trace the horizontal dashed line in Fig. 4).

Figure 5 presents the spin phase diagrams of two-electron NWQDs with respect to the lateral and longitudinal confinements, parametrized by $\hbar\omega_0$ and $\hbar\omega_z$, respectively, in a fixed magnetic field $B = 5$ T for (a)

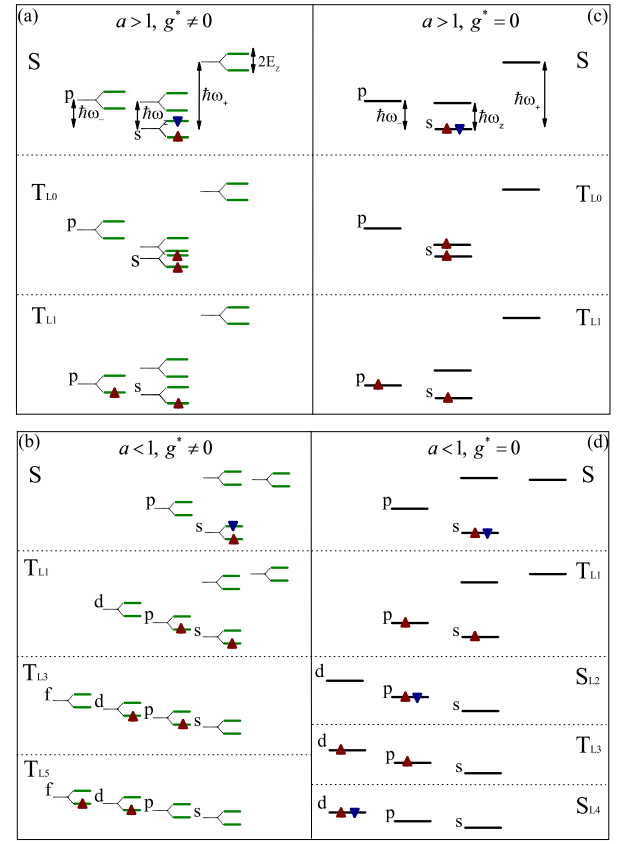
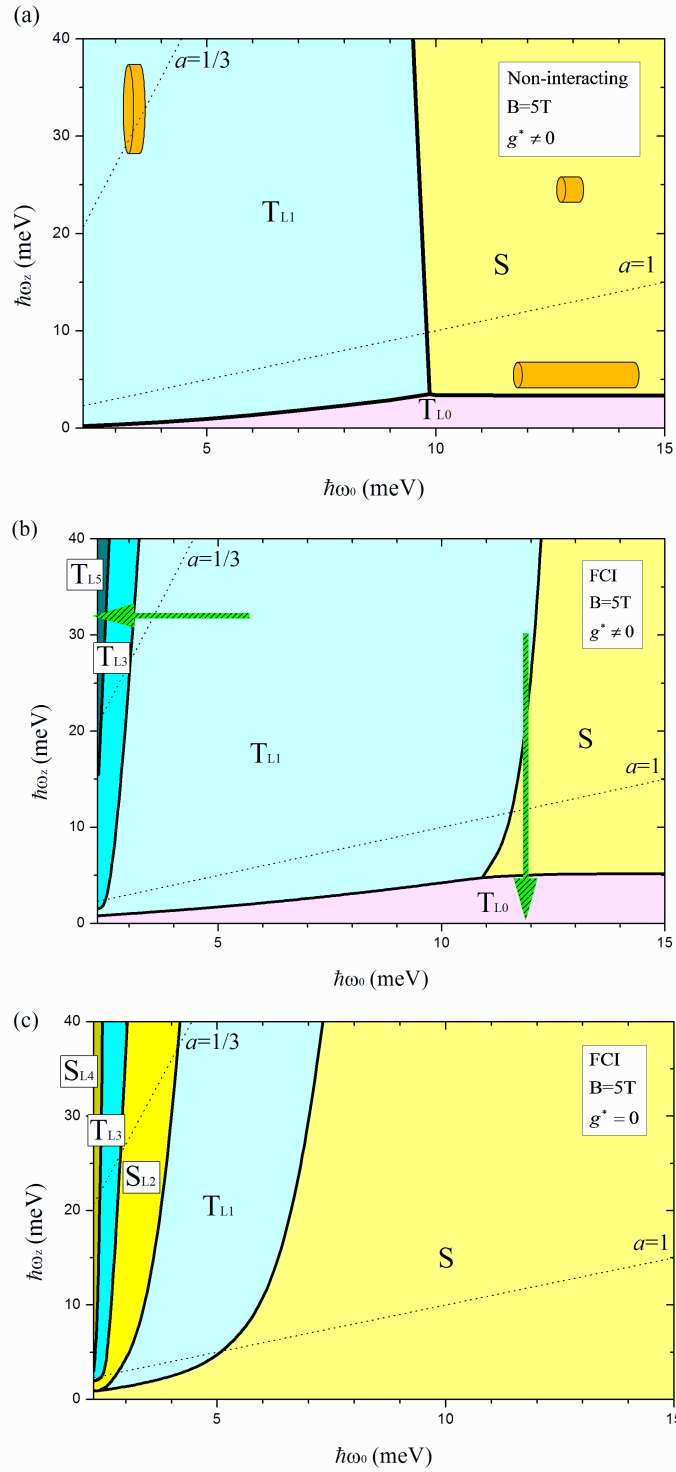


FIG. 6: (Color online) Relevant two-electron configurations possibly being the main components in the ground states of NWQDs in a uniform magnetic field $B = 5$ T for (a) $a > 1$ and $g^* \neq 0$, (b) $a < 1$ and $g^* \neq 0$, (c) $a > 1$ and $g^* = 0$, and (d) $a < 1$ and $g^* = 0$.

non-interacting two electrons with $g^* \neq 0$, (b) interacting two electrons with $g^* \neq 0$, and (c) interacting two electrons with $g^* = 0$. In Figs. 6(a) and (b), we present the relevant two-electron configurations to the spin phase diagrams of Figs. 5(a) and (b) with the inclusion of spin Zeeman effect ($g^* \neq 0$), while in Figs. 6(c) and (d) we present the relevant two-electron configurations to the spin phase diagrams of Fig. 5(c) for $g^* = 0$.

The non-interacting spin phase diagram is first shown in Fig. 5(a) in order to identify the spin Zeeman effect and also contrast the Coulomb interaction effects on the interacting spin phase diagrams presented in Fig. 5(b). In the non-interacting case, the features of the spin phases of Fig. 5(a) are purely determined by the competition between geometry-dependent quantized electronic structures of dots and the spin Zeeman splitting, which is nearly a constant here created by the fixed B . Three distinctive spin phases, S, T_{L0} , and T_{L1} , are marked in different colors in Fig. 5(a). In the yellow region where both $\hbar\omega_0$ ($\hbar\omega_-$) and $\hbar\omega_z$ are large, the kinetic quantizations in both longitudinal and lateral directions are stronger than the spin Zeeman splitting and S remains as a ground state. Reducing the longitudinal confinement,

$\hbar\omega_z$, can lead to the S- T_{L0} (from the yellow to the pink region) transition as $\hbar\omega_z \lesssim 2|E_Z|$. Similarly, reducing the transverse confinement leads to the S- T_{L1} transition as $\hbar\omega_- \lesssim 2E_Z$ (from the yellow to the light cyan region).

Compared with Fig. 5(a), the interacting spin phase diagram of Fig. 5(b) shows the following additional features:

(i) Larger areas of both T_{L0} and T_{L1} phases are observed because of the additional negative exchange energies and the reduced direct Coulomb repulsions gained by the triplet states.

(ii) A NWQD with $\hbar\omega_0 \approx 12$ meV could experience a three-phase transitions from T_{L1} (cyan) to S (yellow), and then to T_{L0} (pink) with increasing the length of wire, from $\hbar\omega_z > 25$ meV to $\hbar\omega_z < 5$ meV (see the vertical line positioned at $\hbar\omega_0 = 12$ meV in Fig. 5(b)).

(iii) In the regime of small $\hbar\omega_0$ and large $\hbar\omega_z$ (i.e. flat quasi-2D dots with $a \ll 1$), a series of transitions from the spin single states to various triplet states, T_{L1} , T_{L3} , T_{L5} , etc. [see Fig. 6(b)] and a staircase increase of total orbital angular momentum are observed with reducing the lateral confinement $\hbar\omega_0$.

In the weak laterally confining regime, few electrons in the quasi-2D QD in a high magnetic field successively fill the orbitals with negative z -projection of orbital angular momentum, i.e. the orbitals of lowest Landau level (LLL), with small kinetic energy separation $\hbar\omega_-$. The inter-particle Coulomb interactions thus become particularly pronounced among the particles on the nearly degenerate LLL orbitals with almost quenched kinetic energies. In order to minimize the coulomb repulsion, the particles on the quasi-degenerate orbitals tend to spread the occupancy of orbitals as far as possible, but in competition with the cost of increase of kinetic energy. As a result, with reducing $\hbar\omega_0$ or increasing B , the total angular momentum of two-electron increases, as previously discussed by Wagner *et al.*⁹ for gated 2D QDs.

Figure 5(c) shows the phase diagram of two interacting electrons calculated by FCI method but with the vanishing spin Zeeman term, i.e. $g^* = 0$. This allows us to distinguish the effects of spin Zeeman energy and the coulomb interactions on the spin phase diagram of Fig. 5(b), and also to study the spin phases of QD made of a material with small g^* such as GaAs. Without spin Zeeman splitting, the significant features of Fig. 5(c) are completely determined by the many-body effects and geometry-engineered electronic structures of NWQDs.

In the $a > 1$ regime, unlike the result shown in Fig. 5(b), the T_{L0} phase disappears and naturally there is no S- T_{L0} transition observed. This is because the Coulomb correlations that energetically favor spin-singlet state as mentioned previously, become dominant and compensate the negative exchange energy gained by the T_{L0}

states.^{6,24} However, in the small $\hbar\omega_0$ regime, an additional singlet-triplet state oscillation with decreasing $\hbar\omega_0$ is observed. Compared with Figure 5(b), the difference is the emergences of various singlet states between the triplet phases. This is due to the removal of spin Zeeman splittings, which energetically favor only the triplet states. Such a singlet-triplet state oscillation is evidenced as a main feature of a flat 2D QD with small spin Zeeman effect, as shown both theoretically⁹ and experimentally¹² in the previous studies.

IV. SUMMARY

In conclusion, we present exact diagonalization studies of spin phase transitions of two electrons confined in nanowire quantum dots with highly tunable aspect ratio and external magnetic field. A configuration interaction theory based on a 3D parabolic model for such three dimensionally confining QDs is developed, which provides generalized explicit formulation of the Coulomb matrix elements and allows for straightforward implementation of direct diagonalization. The exact diagonalization study reveals fruitful features of spin ST transitions with respect to the tunable geometric aspect ratio and applied magnetic field.

For disk-like QDs, the ST transition behaviors may be dominated by the spin Zeeman, the direct-Coulomb, and the exchange energies. The pronounced Coulomb correlations are identified in rod-like QDs with aspect ratio $a > 3$, which energetically favor singlet spin states and yield the always non-zero critical magnetic fields of ST transitions. The developed theory is further employed to study spin phase diagram in the dimensional “cross over” regime from the 2D (disk-like) QDs to finite 1D (rod-like) QDs. In the 2D disk-like QD regime, various distinctive spin phases are emerged under the conditions of appropriate lateral confinement strength and magnetic fields. In the rod-like QD regime, switching the ST transitions is shown feasible by controlling both lateral and/or longitudinal confinement strength.

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